Proofs on Feasibility of Measuring Geologic Age by a Thermal Analysis Method

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Abstract:-We measured the geological age of Mexico Durango apatite, the international standard sample for geological dating, using our previously proposed thermal analysis research approach. Compared with the international recommended age or the calculated age of this sample in this study, the resulting error of the measurement was approximately 1%. This proves the feasibility of measuring geologic age by the proposed thermal analysis method. The dating formula presented in this work to measure the apparent age of Maanshan Anhui apatite directly is easier to implement than other dating methods. Through further research, the precision age of geological sample may be measured with our formula. Compared with fission-track, α ion nuclear track annealing is more sensitive to the temperature of the geological environment; therefore, it might be useful for revealing the thermal history of the geological environment.

Key Words: -Isotopic Dating, Solid State Nuclear Track Detector (SSNTD), Thermal Analysis, DSC Curve, Annealing Heat; Apatite.

1 Introduction

Since fission-track dating was discovered in 1965[1-4] , it has been widely used [5-8], and it remains one of the basic approaches used in isotopic dating[9-17]. However, there are disadvantages to using fission-track dating method. In the fundamental formula for dating,

$$t = \frac{1}{\lambda_D}\ln\left(\frac{\lambda_D N_F}{\lambda_F N_{238}}+1\right) \quad (1)$$

the ^{238}U total decay constant λ_D=1.551×10^{-10}a^{-1}, the spontaneous fission ^{238}U nuclear number N_F, and the existing ^{238}U atom number can be determined accurately, but the numerical value of ^{238}U spontaneous fission constant λ_F has not been unified at present[18].The λ_F value measured with different methods has a great range; there are three λ_F values applied currently: (7.03±0.11)×10^{-17}a^{-1}; 6.9×10^{-17}a^{-1} and 8.46×10^{-17}a^{-1}, and the difference among the higher and lower values is 18.4%. We substituted these two different decay constants into Formula (1) to calculate the age. When the age precision is 10^6, the numerical difference is approximately 18%. However, due to the complex process of chemical etching required to measure N_F, the total uncertainty is obviously larger.

Isotope dating based on apatite (U-Th)/He is a novel technique that has been developed in recent years to avoid measuring λ_F [19-23]. At present, this method has been widely applied in research on shallow crust, dating near-surface material and studying geothermal history in low-temperature areas [22,23]. One of the most important applications of this dating method is the precise measurement of the contents of isotopes, including ^{238}U, ^{232}Th, ^{147}Sm and ^{4}He. The contents of the ^{238}U, ^{232}Th and ^{147}Sm isotopes can be determined by the mature neutron activation analysis method, but the Helium isotope is a gas and is more difficult to measure. Because there is a one-to-one correlation between the Helium isotope formed and α ion nuclear track, we previously proposed a formula for determining geologic age by measuring the α ion nuclear track with thermal analysis instead of determining the ^{4}He isotope formed[24-26].

$$T_0 = \frac{1}{\lambda_D} \ln(1 + \frac{Q_{0\alpha}(x\%)}{8a\overline{E}_\alpha N_{(t)}}) \qquad (2)$$

Using formula (2), in this study, we measure the geological age of Mexico Durango apatite, the international standard sample for geological dating. Compared with the international recommended age or the calculated age in previous studies, the error of our measurement result is approximately 1%. This demonstrates that the method of measuring geologic age by thermal analysis is feasible with the correct formula. In addition, our measurement of the apparent age of apatite collected from Maanshan, Anhui, China, provides a reference for other researchers.

2 Determining Geological Age by the Thermal Analysis Method

In formula (2), the meaning of each parameter in the formula is as follows:

$\lambda_D = 1.551 \times 10^{-10} / a$ refers to the total decay constant (including the sum of spontaneous fission, α decay and β^- decay effect). $Q_{0\alpha}$ refers to the annealing heat in the geological sample unit mass that is measured with thermal analysis, which is produced during α- particle nuclear track annealing and which is released from α decay of all of the isotopes. $x\%$ refers to the ratio of the nuclear track density ρ_α formed by α- particle release from ^{238}U decay to the nuclear track density ρ formed by α- particle nuclear track release from α decay of all of the isotopes (^{238}U， ^{235}U, ^{232}Th and ^{147}S, etc.), namely $x\% = \frac{\rho_\alpha}{\rho}$.□ a refers to the ratio of annealing heat to irradiation dose in the geological sample. $\overline{E_\alpha}$ refers to the average energy of the α- particle release from α decay of all of the isotopes in the geological sample. $N_{(t)}$ refers to the nuclear number of the existing ^{238}U in the geological sample unit mass..

The average values for 238U, 232Th and 147Sm isotopic contents of Durango apatite sample measured by are $30.99\mu g/g$, $265.555\mu g/g$ and $30.99\mu g/g$ [27] , respectively. Thus, it can be calculated that $N_{^{238}U} = 3.443 \times 10^{16} n/g$, $N_{^{235}U} = 2.495 \times 10^{14} n/g$ (^{235}U content is 1/138 of ^{238}U and $N_{^{232}Th} = 6.891 \times 10^{17} n/g$ and

$N_{^{147}S} = 1.269\times10^{17} n/g$. The actual measured value of Helium nucleus is $N_{^4He} = 7.508\times10^{15} n/g$ (calculated with the conversion factor for the Helium content of 1.2472×10^{-8}mol/g[27]). In this study, the age of the Durango apatite sample is calculated to be 30.1Ma. The international recommended age is 31Ma. The α decay constants of ^{238}U, ^{235}U, ^{232}Th and ^{147}Sm isotopes are λ_{238} = 1.537×10^{-10}/ a, λ_{235} = 9.722×10^{-10}/ a, λ_{232} = 0. 499×10^{-10}/ a and λ_{147} = 6.6×10^{-12}/a, respectively. The methods for calculating the parameters in Formula（2）are as follows:

The annealing heat of the α- particle nuclear track released from α decay of all of the isotopes in the Durango apatite sample was measured twice by scanning the DSC curve with a Q2000 differential scanning calorimeter made by TA Instruments, with the scanning speed of℃/min as shown in Figures 1 & 2. This heat can be calculated as follows:

$$Q_{0\alpha} = \frac{1.964+1.914}{2} = 1.939(J/g) \qquad (3)$$

The nuclear track density formed each year from α-particles released from α decay of all the isotopes is:

$$\begin{aligned}\rho &= N_{238}\times8\times\lambda_{238} + N_{235}\times7\times\lambda_{235} \\ &\quad + N_{232}\times6\times\lambda_{232} + N_{147}\times1\times\lambda_{147} \\ &= 3.443\times10^{16} n/g\times8\times1.537\times10^{-10}/a \\ &\quad + 2.495\times10^{14} n/g\times7\times9.722\times10^{-10}/a \\ &= +6.891\times10^{17} n/g\times6\times0.4948\times10^{-10}/a \\ &\quad + 1.269\times10^{17} n/g\times1\times6.6\times10^{-12}/a \\ &= 2494.212\times10^{5} n/ag \end{aligned} \qquad (4)$$

$$\rho_\alpha = N_{238}\times8\times\lambda_{238} = 423.354\times10^{5} n/ag \qquad (5)$$

$$x\% = \frac{\rho_\alpha}{\rho} = 16.973\% \qquad (6)$$

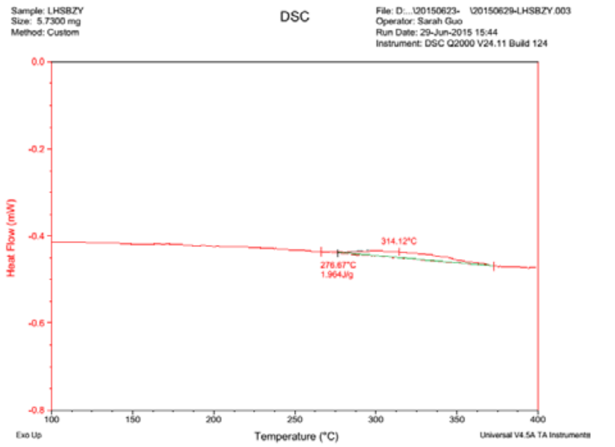

Figure 1 DSC Curve 1 of Durango Apatite Sample

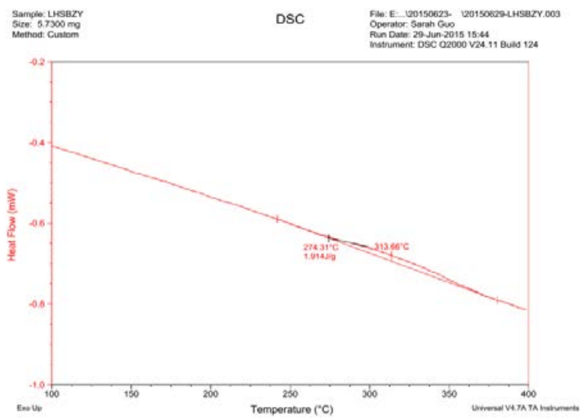

Figure 2 DSC Curve 2 of Durango Apatite Sample

To calculate the average energy of each α-particle after ^{238}U forms the stable isotope ^{206}Pb through 8 α decays:

$$\overline{E_{238}} = \frac{\begin{array}{c}(4.18+4.768+4.658+4.777\\+5.482+6.002+7.6+5.298)MeV\end{array}}{8} \qquad (7)$$
$$= 5.346MeV$$

To calculate the average energy of each α- particle after 235U forms the stable isotope ^{207}Pb through 7 α decays:

$$\overline{E_{235}} = \frac{\begin{array}{c}(4.394+5.046+6.036+5.744\\+6.813+7.356+6.622)MeV\end{array}}{7} \qquad (8)$$
$$= 6.002MeV$$

To calculate the average energy of each α-particle after ^{232}Th forms the stable isotope ^{208}Pb through 6 α decays:

$$\overline{E_{232}} = \frac{(4.007+5.420+5.681 +6.282+6.744+8.785)MeV}{6} = 6.153MeV \quad (9)$$

To calculate the Energy of [147]Sm through 1 α decay:

$$\overline{E_{147}} = 2.23MeV \quad (10)$$

To calculate the average energy released from each α-particle with total α decay of four types of isotopes including [238]U, [235] U, [232]Th, [147] Sm :

$$\overline{E_\alpha} = \frac{N_{238}\times 8\times\lambda_{238}\times\overline{E_{238}} + N_{232}\times 6\times\lambda_{232}\times\overline{E_{232}} + N_{235}\times 7\times\lambda_{235}\times\overline{E_{235}} + N_{147}\times 1\times\lambda_{147}\times\overline{E_{147}}}{N_{238}\times 8\times\lambda_{238} + N_{232}\times 6\times\lambda_{232} + N_{235}\times 7\times\lambda_{235} + N_{147}\times 1\times\lambda_{147}}$$
$$= \frac{(423.354\times 5.346+2045.503\times 6.153 +16.979\times 6.002+8.376\times 2.23)}{2494.418\times 10^5}\times 10^5 = 6.001(MeV) = 9.614\times 10^{-13} J \quad (11)$$

To calculate the ratio of annealing heat to radiation dose:

$$a = \frac{HeatQ_{0\alpha}}{\text{Total number of α particle} \times\text{Average energy of each particle}} = \frac{1.939J/\text{g}}{7.508\times 10^{15} n/g\times 9.614\times 10^{-13} J/n} = 2.686\times 10^{-4} \quad (12)$$

To calculate the existing [238]U nuclear number in the geological sample unit mass:

$$N_{(t)} = 3.443\times 10^{16} n/g \quad (13)$$

Substitute the above values into Formula (2):

$$\mathrm{T} = \frac{1}{\lambda_D}\ln(1+\frac{Q_{0\alpha}(x\%)}{8a\times\overline{E_\alpha}\times N_{(t)}}) = \frac{1}{1.551\times 10^{-10}/a} \times\ln(1+\frac{1.939J/g\times 0.16973}{8\times 2.686\times 10^{-4}\times 9.614\times 10^{-13}\times 3.443\times 10^{16} n/g}) = 2.976\times 10^7 a \quad (14)$$

The error with the calculated age in the study is:

$$\text{Relative error} = \frac{30.1-29.762}{30.1} = 1.124\% \quad (15)$$

By substituting

$N_{^{238}U} = 3.443\times 10^{16} n/g$, $N_{^{235}U} = 2.495\times 10^{14} n/g$ ([235]U content is1/138 of [238]U) , $N_{^{232}Th} = 6.891\times 10^{17} n/g$ and $N_{^{147}S} = 1.269\times 10^{17} n/g$, as well as the recommended age of the Durango apatite standard sample t=31Ma, into the fundamental formula of（U-Th）/He, using the isotopic dating method[21]:

$$N_{^4He} = 8N_{^{238}U}(e^{\lambda_{238}t}-1)+7N_{^{235}U}(e^{\lambda_{235}t}-1) +6N_{^{232}Th}(e^{\lambda_{232}t}-1)+1N_{^{147}S_m}(e^{\lambda_{147}t}-1) \quad (16)$$

then $N_{^4He} = 7.754\times 10^{15} n/g$ can be calculated, which is the theoretical value based on the recommended age of the Durango apatite standard sample. Substituting this value into Formula （12）, allows $a=2.601\times 10^{-4}$ to be calculated, and substituting this value into Formula (14), allows the geological age of Dorango apatite $T=3.073\times 10^7$ to be calculated and compared with the international recommended age.

$$\text{Relative error} = \frac{31-3.073}{31} = 0.862\% \quad (17)$$

Compared with the international recommended age or the calculated age of this sample in this study, the error is approximately 1%. This result suggests that the scheme of measuring geological age by the proposed thermal analysis method is feasible with the correct formula. The geological age of one sample will be tested below with this method and formula.

We scanned the DSC curve of apatite collected from Maanshan with a Q2000 differential scanning calorimeter made by TA Instruments with the scanning speed of 5℃/min. shown in Figures 3 & 4. The results were calculated by the following formula:

$$Q_{0\alpha} = \frac{2.102 + 2.019}{2} J/g = 2.061(J/g) \quad (18)$$

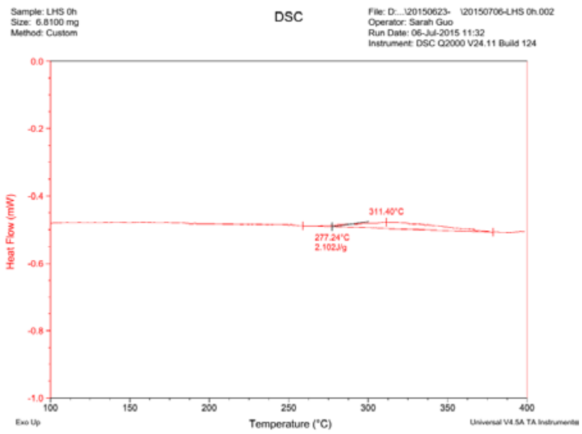

Figure 3 DSC Curve 1 of the Maanshan Apatite

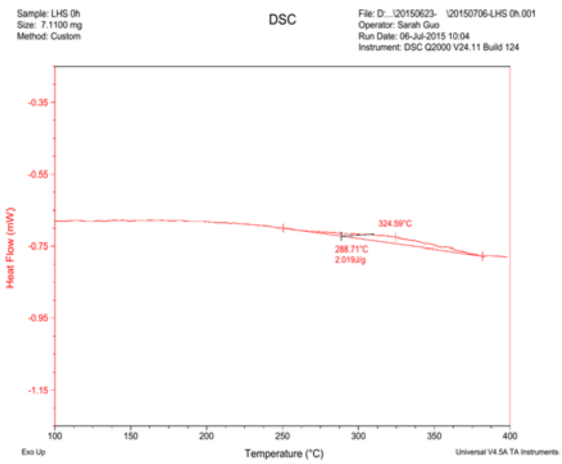

Figure 4 DSC Curve 2 of the Maanshan Apatite

Neutron activation analysis was conducted by the China Institute of Atomic Energy, and the uranium and thorium contents of this sample are shown in Table 1.

Table 1. Neutron Activation Analysis of the Geological Sample

Sample Name	Quality (mg)	U Content	Th Content	Sm
Maanshan Apatite	100.5	26.2 ± 2.9%ppm	189.6 ± 2.2%ppm	0

The following result can be calculated using the data in Table 1:

$$N_{^{238}U} = 6.627 \times 10^{16} n/g \quad (19)$$

$$N_{^{235}U} = 4.800 \times 10^{14} n/g \quad (20)$$

$$N_{^{232}Th} = 4.920 \times 10^{17} n/g \quad (21)$$

$$N_{^{147}Sm} = 0 . \quad (22)$$

$$x\% = \frac{\rho_a}{\rho} = \frac{8N_{238}\lambda_{238}}{8N_{238}\lambda_{238} + 7N_{235}\lambda_{235} + 6N_{232}\lambda_{232}} = 0.363 \quad (23)$$

(24)

$$\overline{E_\alpha} = \frac{N_{238} \times 8 \times \lambda_{238} \times \overline{E_{238}} + N_{232} \times 6 \times \lambda_{232} \times \overline{E_{232}} + N_{235} \times 7 \times \lambda_{235} \times \overline{E_{235}}}{N_{238} \times 8 \times \lambda_{238} + N_{232} \times 6 \times \lambda_{232} + N_{235} \times 7 \times \lambda_{235}} = 5.862 MeV = 9.391 \times 10^{-13} J$$

The ratio a of annealing heat to irradiation dose can be tested through laboratory simulation. We used the α- particle released from a ^{239}Pu radioactive source to irradiate the annealed Maanshan apatite sample and measured the annealing heat Q with differential scanning calorimeter after a period of irradiation time. Then, we calculated the irradiation dose G according to the strength of the activity of radioactive source, the amount of particle energy released, the quality of the irradiated sample and the irradiation time. Thus, the ratio a of annealing heat to irradiation dose can be calculated as follows:

$$a = \frac{Q}{G} = 0.02315 \quad (25)$$

These parameters may be substituted into

Formula (2):

$$T = \frac{1}{\lambda_D}\ln(1+\frac{Q_{0\alpha}(x\%)}{8a\times\overline{E_\alpha}\times N_{(t)u_{238}}}) \\ = \frac{1}{1.551\times10^{-10}/a} \\ \times\ln(1+\frac{2.0605J/g\times0.3631}{8\times0.02315\times9.391\times10^{-13}\times6.627\times10^{16}n/g}) \\ = 4.185\times10^{5}a \quad (26)$$

In Formula (25), Q is the annealing heat of theα- particle nuclear track produced by a radioactive source. Here, the nuclear track in the sample does not consider the effect of the annealing factor in the geological environment; therefore, the measured age is younger than the actual age of the sample, while the precise age can be determined only after correction.

3. Discussion

From our calculations , we found that the irradiation dose for the sample with αdecay of ^{238}U, 2^{32}T and $^{147}S_m$ isotopes is:

$$G = N_{^4He}\times\overline{E_\alpha} = 7.508\times10^{15}(n/g)\times6.001MeV/n \\ = 7218.5071J/g \quad (27)$$

From the measured annealing heat value of the Maanshan apatite sample, it can be determined that its irradiation dose will not be less than the above value. Obviously, the α- particle irradiation dose for the general geological sample is considerable. The general process of irradiation energy loss occurs in the following three ways: When the α- particle energy is high, electromagnetic radiation is produced because of an abrupt deceleration of the particles in the target nucleus; this is Bremsstrahlung energy loss. When the α- particles interact with electrons, an instantaneous state of disorder and a semi-permanent effect can be produced in the crystal for only a short time (less than 10^3 s). Meanwhile, when the α- particles collide with atoms, destroying the crystal structure, the displaced atoms formed via this collision can last a long time in the crystal, causing permanent damage and producing nuclear tracks[28-30].Thermal analysis instruments are used to measure the energy released from lattice recovery when heating and annealing. The sensitivity of thermal analysis instruments has reached μW (The sensitivity of Q2000 made by TA Instruments is 0.2μW), even nW order of magnitude, so there is no difficulty in measuring the heat in reality. At present, the heat collected by thermal analysis instruments accounts for 20%—97% of the total heat released by the samples. This suggests that the measured heat will be different when using a different instrument or a different method on the same instrument. In practice, if the annealing heat of a natural geological sample and a synthetic sample radiated by an irradiation source are measured under the same conditions (the same instruments, measurement methods, sample processing and even instrument operators), the error caused by measuring annealing heat with different instruments or by different methods with the same instrument will be reduced. According to the mechanism of forming displaced atoms, the energies of the α- particles are mainly below 1 MeV when forming displaced atoms; therefore, the effect of the selected α- particle energy can be ignored.

The effect of the annealing behavior of nuclear tracks in the geological environment relative to the measured apparent age must be studied for all isotopic dating methods so that the real age can be obtained through correcting the apparent age. Compared with the fission track, it is easier for the nuclear track of the α- particle to be affected by the annealing behavior in the geological environment. Therefore, studying the annealing behaviors of the nuclear tracks from α- particles in geological environments is important. The age of the geological sample can be measured accurately and precisely using the thermal analysis dating method after the annealing behavior in the geological environment has been determined.

4 Conclusion

Apatite (U-Th)/He isotope dating via thermal analysis was used in a detailed geological dating study to determine the apparent age of Maanshan Anhui apatite directly, revealing an apparent age of $1.8782\times10^{6}a$. This proves the feasibility of geologic dating using our previous formula $T_0=\frac{1}{\lambda_D}\ln(1+\frac{Q_{0\alpha}(x\%)}{8a\bar{\bar{E}}_\alpha N_{(t)}})$. Revealing the thermal history of the geological environment might be advantageous because α ion nuclear track annealing is sensitive to the temperature of the geological environment and easy to measure.

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